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Calibration of nuclear track detectors for the measurement of indoor radon and thoron levels

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Abstract : Solid State Nuclear Track Detectors are increasingly being used to obtain the time integrated concentration levels of radon/thoron and their daughters. It is essential that these detectors should be properly calibrated and standardised for such applications. In this paper the procedure for the calibration of LR-115 type-II and CR-39 track detectors are presented. The detectors were exposed to different concentration of radon/thoron alongwith monodisperse aerosol of 0.2 μm aerodynamic size in the exposure chamber The estimated sensitivity factors for the LR-115 type-II for radon and its daughters are 0.0284 tracks $cm^{-2}.d^{-1}$ per $Bq.m^{-3}$ and 490.0 tracks. $cm^{-2}.d^{-1}$ per WL. The sensitivity factors for the CR-39 detectors for radon and its daughters are 0.182 tracks. $cm^{-2}.d^{-1}$. per $Bq.m^{-3}$ and 1628.0 tracks.$cm^{-2}.d^{-1}$ per WL. The sensitivity factor for LR-115 type-II for thoron and its daughters are 0 538 tracks.$cm^{-2}.d^{-1}$ per $Bq.m^{-3}$ and 0.317 tracks.$cm^{-2}.d^{-1}$ perm WL. The sensitivity factors for radon obtained in this work are in good agreement with other published results.

Keywords : Nuclear track detectors, calibration, indoor radon, thoron, aerosol, sensitivity factors.

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1. Introduction

There has been an increasing interest in indoor radioactivity measurements motivated by the concern about the possible consequences of long-term exposure to higher concentration of radon-222 and radon-220 and their progeny. Eventhough radon-220 (thoron) and its progeny are also an important component of the natural radiation environment, relatively less attention has been given to them in the indoor radiation environmental studies due to their low concentrations. But in certain locations, high concentrations of thoron and its progeny may be of great concern (Paul *et al* 1982).

Several techniques are in use to measure the radon/thoron and their daughter products in air. This includes their collection on a filter paper and subsequent alpha counting using ZnS (Ag) surface barrier detectors or ionizations chambers. Several personnel dosimeters employing track-etch detectors, TLD (Thermo Luminicent

Detector) and photographic films have also been developed. These techniques have been used in the time integrated mode for the measurement of radon and thoron in the soil gas for uranium exploration, earth-quake prediction and geological and geophysical studies.

The track-etch detectors used in the present work for the measurement of thoron/radon and its progeny in the environment are cellulose nitrate films, available commercially as the red dyed Kodak LR-115 type-II films of 13 μm thick from Kodak Path, France, and the CR-39 polycarbonate foils (250 μm thick) from Pershore Moulding, UK. The basic properties of these detector foils have been given in detail elsewhere (Ramachandran et *al* 1987b, 1990). The measurement technique involves exposing a small strip of the above detectors to the atmosphere to be monitored. Alpha particles from the daughters of radon/thoron or both, present in the air, bombard the plastic films and produce tracks due to radiation damage, which are subsequently, revealed by caustic soda etching and are counted using a microscope at suitable magnification. Number of tracks recorded per unit area ($T.cm^{-2}$) is proportional to the average exposure rate ($Bq.m^{-3}$) and the exposure time (t). The exposure time can vary from a few days to few weeks depending on the concentrations encountered.

2. Standardisation procedure

In order to evaluate the concentration in terms of the activity of radon/thoron and their daughters present in the air, the detector has to be calibrated using known concentrations of radon/thoron and their daughter products. Calibration of the detectors takes into consideration the following features : (a) accurately known radon/thoron level ; (b) well characterised environment with respect to daughter equilibrium, aerosol size, concentration and humidity ; (c) instrumentation for monitoring radon/thoron and their daughter's concentrations and (d) uniformity of radon/thoron content in the calibration chamber.

In the calibration method employed, two detector configurations were used : (a) "Bare mode" and (b) "Filter cup or Membrane cup mode". In the "Bare mode", the detector foils of 2.5 cm $\times$ 2.5 cm size is mounted flat on a rectangular card such that it views a hemisphere of air of radius at least 9.1 cm, the range of a Po-212 alpha in air, or 6.9 cm, the range of the Po-214 alpha (if only the radon-222 decay series is present). No surface should be closer than this range as daughter deposition would then add an indeterminate alpha particle source to be registered on the detector. The Track Etch reading of a "Bare" detector will be a function not only of radon/thoron, but of the degree of equilibrium of radon with its daughters. The "Bare" detector mode of exposure is therefore a measure of the total potential alpha energy exposure (PAEC) expressed in Working Level (WL) units.

The membrane cup mode consists of a plastic cup fitted with a detector of 2.5 cm × 2.5 cm size at the bottom of the cup. The open mouth of the cup is covered with a semipermeable membrane. This membrane, allows the normal diffusion of noble gases into the cup thus discriminating radon from thoron, while permitting 60 to 70% of Radon-222 only (Ramachandran et *al* 1987a). This configuration is generally used in exploration to eliminate thoron interference and water condensation. The membrane cup configuration also prevents the entrance of radon daughters and is a radon-only device.

In the filter cup mode, the open mouth of the cup is covered with a hydrophobic microporous filter which permits complete infiltration of radon isotopes but discriminates against the non-gaseous radon daughters. Because of its higher sensitivity to radon, this configuration is preferred to the membrane in all radon-only applications where thoron is not an important component.

The calibration system consists of an exposure chamber ; an aerosol generator ; a radon/thoron gas generator ; chemical etching equipments and a microscope ; radon/thoron gas collection chambers, conventional filter paper sampler and alpha counting unit. Figure 1 gives the block diagram of the set up used for the calibration

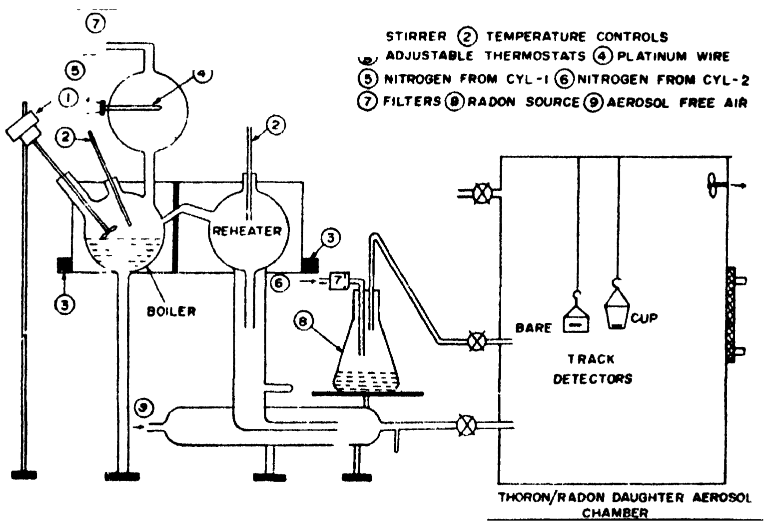

Figure I. Schematic diagram of the apparatus used for the calibration of track detectors.

of the track detectors. The exposure chamber is a painted wooden box of 0.5 m^3 capacity with a couple of inlet ports and an outlet tube and a viewing glass pannel. There is an air-tight window for introducing filtered air, aerosols, radon/thoron gas into the chamber. Outlet tube discharges the contents outside the laboratory.

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The aerosol generator used was a LaMer-Sinclair type condensation aerosol generator (Subba Ramu 1978). This gives a laminar flow of monodisperse aerosol of di-2-ethyl hexyl sebacate condensed on NaCl nuclei. Temperature settings of the re-heater and the boiler could be adjusted in such a way to obtain monodisperse aerosols of 0.2 μm aerodynamic size. 0.2 μm has been chosen for this study since AMAD (Activity Median Aerodynamic Diameter) value for indoor radon daughters is estimated to be 0.2 μm (Muraleedharan et *al* 1986).

The calibration procedure consists of providing a continuous source of radon/thoron gas in the chamber where a known concentration of aerosol was present. The radioactive radon/thoron daughters, which are "solid" atoms get attached to the aerosols. The word "solid" is used as an adjective in order to differentiate the radon daughter atom from any other liquid or gaseous atom. Also since they are solid atoms, they easily attach to aerosols. Radon source used for the experiment was a solution of 1.85×10^5 Bq. of Ra-226 (in the form of radium nitrate) in 150 ml of 1.0 N HNO_3 kept in the flask. For thoron, thorium nitrate packed in small bags were used. The track detectors were exposed to the environment for a known period of time and were chemically etched and scanned under an optical microscope. The radon/thoron/daughter concentrations were maintained uniform through-out in the chamber with the help of a few thousand particles per cm^{-3}. The aerosol chosen for the present experimental work were found to be quite stable during the experimental period. Both diffusional and deposition losses where minimum for the monodisperse aerosols size of 0.2 μm used for the present experiments.

Radon/thoron and its daughter activity levels during the exposure of the detectors were estimated from the activity collected on a millipore filter paper type AA by drawing a known volume of air (50 litre) from the chamber using a vacuum pump and then following the alpha decay of the filter paper sample using an alpha counting unit upto 180 minutes at different time intervals. From the alpha counts, the activity levels of the daughters as well as the radon/thoron gas were estimated with the help of a computer programme (Rangarajan and Datta 1976) incorporating necessary steps to evaluate the wall deposition rate, radon/thoron concentration and the radon/thoron potential alpha energy exposure in terms of working level (WL). One WL is a measure of the total potential alpha energy exposure of radon/thoron in one litre of air which will result in the ultimate emission by them of 1.3×10^5 MeV of alpha-ray energy.

The radon and thoron working level (WL) concentration were estimated from the relation :

$$\text{Radon (WL)} = 2.78 \times 10^{-5}(\text{Ra A}) + 1.37 \times 10^{-4}(\text{Ra B}) + 1.01 \times 10^{-4}\ (\text{Ra C})$$

where Ra A, Ra B and Ra C are the concentration of radon daughters in $Bq.m^{-3}$.

$$\text{Thoron (WL)} = 3.3 \times 10^{8} \text{ (Th B)} + 3.35 \times 10^{-4} \text{ (Th C)}$$

where Th B and Th C are the thoron daughter concentrations in $Bq.m^{-3}$.

During the course of each set of experiments on an average 5 to 7 air filter samples were collected at different time intervals and were analysed for radon/thoron and their daughter concentrations and the average is taken as the radon/thoron and radon/thoron WL concentration for the particular set of the experiment. Relative humidity inside the chamber was about 80%.

After the exposure period, the films were etched by treating them in an alkali solution of 2.5 N NaOH at a constant temperature of 60°C, for a period of 100 minutes for LR-115 and 6 N KOH at 60°C for 6 hours for CR-39. The etching conditions were set for optimum results, in a series of preliminary standardisation experiments carried out earlier (Ramachandran *et al* 1986).

3. Results and discussion

The experiments were carried out to evaluate the relationship between the track density recorded and the radon/thoron concentration as well as the Working Level Concentration. Table 1 gives the sensitivity factors obtained for LR-115 type-II detectors exposed in the "cup with membrane" and "Bare" modes for radon and the potential alpha energy (WL) concentrations. The radon concentration varies from 92.5 to 2146.0 $Bq.m^{-3}$ and the measured track density varies from 2.4 to 60.0 tracks. $cm^{-2}.d^{-1}$. The WL concentration varies from 0.01 to 0.39 WL and the corresponding track density varies from 3.2 to 250.0 tracks.$cm^{-2}.d^{-1}$. From the table, it is seen that the average sensitivity factor for the LR-115 type-II detectors exposed in the "cup with membrane" and "Bare modes" are 0.0284 tracks. $cm^{-2}.d^{-1}$ per $Bq.m^{-3}$ with a standard deviation of 0.0033 and a relative standard deviation of 11.5% for radon and 490.0 track.$cm^{-2}.d^{-1}$ per WL with a standard deviation of 81.0 and a relative standard deviation of 16.8%.

Table 2 gives the sensitivity factors for the CR-39 detectors exposed in the "cup with membrane" and "Bare mode" for radon and the potential alpha energy (WL) concentrations. The radon concentration varies from 251.0 to 2549.0 $Bq.m^{-3}$ and the measured track density varies from 51.0 to 399.0 track.$cm^{-2}.d^{-1}$ for radon with an average of 0.182 track. $cm^{-2}.d^{-1}$ per $Bq.m^{-3}$ and with a standard deviation of 0.023 and relative standard deviation of 12.8%. The WL concentration varies from 0.042 to 0.512 and the measured track density varies from 90.0 to 615 tracks.$cm^{-2}.d^{-1}$. The average sensitivity factor is 1628 track. $cm^{-2}.d^{-1}$ per WL with a standard deviation of 392.0 and a relative standard deviation of 24.1%.

Table 3 gives the sensitivity factor for the LR-115 type-II detectors exposed in the "cup with filter" and "Bare" mode for Thoron and WL concentration. The radon concentration varies from 12.0 to 205.0 $Bq.m^{-3}$ and the measured track density varies from 5.0 to 118.0 $track.cm^{-2}.d^{-1}$. The average sensitivity factor is 0.538 track. $cm^{-2}.d^{-1}$ per $Bq.m^{-3}$ with a standard deviation of 0.17 and a relative standard deviation of 30.8%. The thoron WL concentration varies from 37.0 to 648 mWL and the measured track density from 10.3 to 174.0 $track.cm^{-2}.d^{-1}$. The average

Table I. Calibration factor for the LR-115 type-II detectors exposed in the Cup with Membrane and Bare mode for radon and potential alpha energy concentration (WL).

Concentration of radon ($Bq.m^{-3}$)	Measured track density ($T.cm^{-2}.d^{-1}$)	Sensitivity ($T.cm^{-2}.d^{-1}$ per $Bq.m^{-3}$)	Potential alpha energy concentration (WL)	Measured track density ($T.cm^{-2}.d^{-1}$)	Sensitivity ($T.cm^{-2}.d^{-1}$ per WL)
92.5	2.4	0.0260	0.0100	3.2	320.0
136.9	3.3	0.0241	0.0110	3.9	355.0
151.1	4.5	0.0283	0.0225	7.9	351.0
185.0	5.1	0.0276	0.0330	13.0	394.0
266.4	7.4	0.0278	0.0520	24.0	462.0
277.5	7.6	0.0274	0.0760	35.0	461.0
314.5	7.6	0.0242	0.1150	53.0	461.0
351.5	12.0	0.0341	0.1400	68.0	486.0
518.0	17.0	0.0328	0.2200	130.0	591.0
703.0	24.0	0.0341	0.2700	160.0	593.0
925.0	25.0	0.0270	0.3200	200.0	625.0
971.0	28.0	0.0280	0.3500	220.0	629.0
2146.0	60 0	0.0280	0.3900	250.0	641.0
Average sensitivity = 0.0284					490.0
S.D. = 0.0033					81.0
R.S.D. = 11.5%					16.8%

sensitivity factor work to be 0.317 $tracks.cm^{-2}.d^{-1}$ per mWL with a standard deviation of 0.067 and a relative standard deviation of 21.0%.

The above type of track-etch detector materials (LR-115) have also been recently calibrated as a part of Asian/Australiasion Regional Inter-Comparison program for radon carried out by Australian Radiation Laboratory, Melborne, Australia during September-November, 1987 and our results have been found to be within ± 10% of the absolute value.

Table 4 presents some of the reported sensitivity factors alongwith the results from the present study. From this table, it is clearly seen that our sensitivity

Table 2. Calibration factor for the CR-39 detector exposed in the Cup with Membrane and Bare mode for radon and potential alpha energy concentration (WL).

Radon concentration ($Bq.m^{-3}$)	Measured track density ($T.cm^{-2}.d^{-1}$)	Sensitivity ($T.cm^{-2}.d^{-1}$ per $Bq.m^{-3}$)	Potential alpha energy concentration (WL)	Measured track density ($T.cm^{-2}.d^{-1}$)	Sensitivity ($T.cm^{-2}.d^{-1}$ per WL)
251.0	50.9	0.205	0.042	89.8	2138.0
299.1	57.3	0.197	0.055	121.5	2211.0
321.5	67.6	0.210	0.058	92.4	1594.0
353.3	68.8	0.195	0.062	108.2	1746.0
484.7	90.4	0.187	0.111	166.6	1501.0
1069.2	188.9	0.177	0.191	354.2	1854.0
1553.8	259.9	0.165	0.399	502.5	1260.0
2008.9	288.0	0.143	0.494	566.6	1147.0
2549.0	399.0	0.156	0.512	614.6	1201.0
Average sensitivity = 0.182					1628.0
S.D. = 0.023					392.0
R.S.D. = 12.8%					24.1%

Table 3. Calibration factor for LR-115 type-II detectors exposed in the Cup with Filter and Bare mode for thoron and potential alpha energy concentration (mWL)*

Thoron concentration ($Bq.m^{-3}$)	Measured track density ($T.cm^{-2}.d^{-1}$)	Sensitivity ($T.cm^{-2}.d^{-1}$ per $Bq.m^{-3}$)	Potential alpha energy concentration (mWL)	Measured track density ($T.cm^{-2}.d^{-1}$)	Sensitivity ($T.cm^{-2}.d^{-1}$ per mWL)
12.0	5.0	0.416	36.55	12.95	0.354
14.8	6.3	0.422	36.91	10.29	0.279
27.4	9.4	0.343	84.91	17.52	0.206
33.1	10.5	0.316	108.62	41.89	0.386
51.4	31.6	0.615	173.75	69.12	0.398
57.4	41.6	0.724	389.48	112.32	0.288
109.0	76.9	0.706	466.31	149.38	0.320
131.1	95.3	0.727	648.19	173.89	0.268
205.1	118.2	0.577	—	—	–
Average sensitivity = 0.538					0.317
S.D. = 0.17					0.067
R.S.D. = 30.8%					21.1%

*mWL = milli Working Level.

factors for radon are in agreement with those reported by others. However, there is no reported value for the sensitivity factor for the WL estimation and thoron concentration. Use of typical aerosols representative of those present in a dwelling for the calibration of "Bare" detectors to measure the WL concentration has resulted in obtaining a reasonably accurate calibration factors for WL measurements.

Table 4. Comparison of reported sensitivity factor with the present value.

Reference	Type of detector	Source	Sensitivity factor $\frac{(\mathrm{T.cm^{-2}.d^{-1}})}{(\mathrm{Bq.m^{-3}})}$	Sensitivity factor $\frac{(\mathrm{T.cm^{-2}.d^{-1}})}{(\mathrm{WL})}$	Remarks on detector placement
Alter and Fleischer (1981)	Terradex	Radon	6.03×10^{-2}	-	Placed in a plastic cup covered with a semi-permeable membrane exposed in an experimental chamber
Savage (1983)	,,	,,	5.65×10^{-2}	--	Placed in a cup with membrane and exposed in a chamber
Giridhar and Raghavayya (1986)	LR-115	,,	2.00×10^{-2}	—	Placed in a plastic cup with special rubber membrane and exposed in a chamber
Chrusciele Wski *et al* (1982)	LR-115	,,	–	4.2×10^{2}	Exposed in a bare mode to mine air and a simulation chamber and for a F 0.25
Urban *et al* (1985)	CR-39	,,	0.17	—	Placed in diffusion chamber (cup) with a hydrophobic fiber glassfilter
Present	LR-115	,,	2.84×10^{-2}	4.89×10^{2}	Aerosol load provided to simulate dwelling condition
,,	CR-39	,,	0.18	1628.0	,,
,,	LR-115	Thoron	0.538	0.3170 (per mWL)	,,

These calibration results are very useful in planning the application of nuclear track detector method for the monitoring of indoor and environmental radon/thoron and its daughter levels. The detector foils are found to be suitable and easy to be used for exposure both in the "Bare" and in the "cup with membrane" and "cup with filter" mode. In our calibration experiments, we have used aerosols of known AMAD to simulate the atmospheric conditions of a dwelling. The thoron and radon concentration levels used in the calibration experiments varied from low to high values and are higher than the levels normally prevalent in the indoor environment (Mishra and Subba Ramu 1987, Ramachandran *et al* 1984, 1989). However, it can be seen that the calibration can be linearly extrapolated down to the lower levels.

4. Conclusion

Based on the calibration experiments, some of the main conclusions drawn are : (a) The system used for the calibration of LR-115 and CR-39 detectors has been found to be quite convenient and accurate enough to obtain the conversion factor between the tracks per unit area and the concentration of radon/thoron in $Bq.m^{-3}$ or WL for a given period of exposure of the film either in the "Bare mode" or in the "cup with membrane" or "cup with filter mode". (b) Use of typical aerosols of the same characteristics as one normally found in a dwelling or an enclosed area or room has facilitated the calibration of the "Bare" detector mode to estimate the WL concentration within $\pm$ 17% to 25%. (c) The concentrations of radon, thoron and their daughters used in the calibration experiments are more than those normally encountered in dwellings or rooms except in some uranium or non-uranium and high background areas. However, the calibration graph can be extrapolated and be used for the measurements of low concentration too. It is important to point out here that the statistics of track counting must be improved by both increasing the number of days of exposure and covering a large area of the exposed films while counting. (d) The sensitivity factors estimated for the LR-115 type-II foils for radon are 0.0284 $track.cm^{-2}.d^{-1}$ per $Bq.m^{-3}$ and 490,0 $track. cm^{-2}$ per WL ; for the CR-39 foils for radon are 0.18 $track. cm^{-2}.d^{-1}$ per $Bq.m^{-3}$ and 1628 $track.cm^{-2}.d^{-1}$ per WL ; for the LR-115 type-II for thoron are 0.538 tracks. $cm^{-2}.d^{-1}$ per $Bq.m^{-3}$ and 0.317 $track.cm^{-2}.d^{-1}$ perm WL. These factors are relative to the exposure conditions used in this experiment.

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